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14. ABSTRACT
Approximately fifteen years ago the AFOSR recognized the potential of using ultrafast lasers to study chemical reactions and funded Professor Zewail in his endeavor to unravel the essential chemical processes that occur during chemical reactions. In 1999, Professor Ahmed H. Zewail received the Nobel Prize in Chemistry for the development of Femtochemistry. At the 220th ACS National Meeting in Washington a symposium has been organized to honor Professor Zewail's accomplishments with two goals. The first one is to honor Professor Zewail. The second is to get a cross section of the present state of research in the field of Femtochemistry. The open interchange of ideas is very valuable in this highly technical scientific endeavor. AFOSR funds were requested to defray the cost of travel for the attending speakers. The conference included a number of poster presentations, scientific talks and a plenary talk by Professor Zewail.

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Femtochemistry, Nobel, Symposium, ACS National Meeting

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Final Conference Report
Femtochemistry: 1999 Nobel Prize Symposium
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Approximately fifteen years ago the AFOSR recognized the potential of using ultrafast lasers to study chemical reactions and funded Professor Zewail in his endeavor to unravel the essential chemical processes that occur during chemical reactions. In 1999, Professor Ahmed H. Zewail received the Nobel Prize in Chemistry for the development of Femtochemistry.

At the 220th ACS National Meeting in Washington a symposium was organized to honor Professor Zewail's accomplishments with two goals. The first one was to honor Professor Zewail. The second was to get a cross section of the present state of research in the field of Femtochemistry. The open interchange of ideas is very valuable in this highly technical scientific endeavor.

AFOSR funds were requested to defray the cost of travel for the attending speakers. The conference included a number of poster presentations, scientific talks and a plenary talk by Professor Zewail.

The symposium was a tremendous success. The afternoon hall could sit 500 attendees. The turnout was so great that hundreds of attendees were standing during Zewail's lecture.

Clearly all the goals of this symposium were met.

Please let me know if additional information is required.

Best Regards,

Marcos Dantus
Assistant Professor

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Schedule of Oral Contributions

Femtochemistry: 1999 Nobel Prize Symposium
Honoring Professor Ahmed H. Zewail, Chemistry Nobel Laureate
The 220th ACS National Meeting, Washington DC. 2000
August 24, 2000
Session I

M. Dantus, Organizer

M. Chergui Sr., Presiding

8:00 — 542. Femtosecond techniques to monitor and control chemical reactions on isolated molecules. T. Baumert

8:40 — 543. Some interesting observed properties of metals confined in time and space of different shapes. M. A. El-Sayed

9:20 — 544. Ultrafast interferometry studies of light localization in photonic structures. Y. Liau, N. F. Scherer

10:00 — Coffee Break

10:20 — 545. Time-dependent electronic density matrix simulations of nonadiabatic photodynamics in molecules and aggregates. S. Mukamel, M. Tommasini, V. Chernyak, C. Scheurer

11:00 — 546. Time-resolved dynamics in anion clusters using femtosecond photoelectron spectroscopy. D. M. Neumark

11:20 — 547. Femtosecond energy-transfer dynamics in iron-sulfur proteins. D. Zhong, A. H. Zewail

11:40 — 548. Dynamic stark shifting of molecular energy levels by femtosecond laser pulses. R. B. Lopez-Martens, T. W. Schmidt, G. Roberts

Sponsors: AFOSR, Positive Light, Spectra-Physics Lasers

Schedule of Oral Contributions

Femtochemistry: 1999 Nobel Prize Symposium
Honoring Professor Ahmed H. Zewail, Chemistry Nobel Laureate
The 220th ACS National Meeting, Washington DC. 2000
Session II

M. Dantus, Organizer

M. A. El-Sayed, Presiding

1:00 — 581. Femtochemistry: Some New Directions. A. H. Zewail

1:40 — 582. Probing wavepacket dynamics with femtosecond energy- and angle-resolved photoelectron spectroscopy. K. Takatsuka, Y. Arasaki, K. Wang, V. McKoy

2:20 — 583. Influence of hydration on the dynamics of proton transfer reactions. A. W. Castleman Jr., D. E. Folmer, E. S. Wisniewski, D. A. Card, S. M. Hurley

3:00 — Coffee Break

3:20 — 584. Extracting molecular interactions from observations of controlled quantum dynamics phenomena. H. Rabitz

4:00 — 585. Initiating protein folding at the transition state. M. Gruebele

4:40 — 586. Spin-orbit relaxation and recombination dynamics in I2- and ICl- cluster ions: A new type of photofragment caging reaction. W. C. Lineberger, T. Sanford

5:00 — 587. Ultrafast observation and control of molecular dynamics: Beyond the pump-probe method. M. Dantus, I. Pastirk, V. V. Lozovoy, B. I. Grimberg

Sponsors: AFOSR, Positive Light, Spectra-Physics Lasers

Summary of Scientific Papers Presented

Femtochemistry: 1999 Nobel Prize Symposium
Honoring Professor Ahmed H. Zewail, Chemistry Nobel Laureate
The 220[th] ACS National Meeting, Washington DC. 2000
August 24, 2000

1. **PHYS433** Different surface modifications methods to allow electron-hole recombination in semiconductor nanoparticles Clemens Burda, Stephan Link, Reginald B. Little, Mona Mohamed, and Mostafa A. El-Sayed, School of Chemistry and Biochemistry, Georgia Institute of Technology, Atlanta, GA 30332-0400, Fax: 404-894-7452, burda@chemistry.gatech.edu As the size of matter decreases and becomes comparable to or smaller than the length scale of the electron-hole Bohr radius in a semiconductor material (nanometers), the ratio of the surface to bulk atoms increases, which leads to an enhancement of the surface-state properties. Consequently, the charge carrier dynamics of nanomaterials exhibit size dependencies, which are mainly due to the two main contributions quantum size effect and influence of the interface. The trapping of the electron and interfacial electron transfer was studied in various systems with femtosecond time-resolved transient spectroscopy. By changing systematically the surface of CdE (E=S, Se) nanoparticles we found a number of interesting effects which lead to changes of the relaxation dynamics. The addition of large band-gap capping materials such as ZnSand OH-layers lead to removal of trapping sites and therefore extent the exciton lifetime. The addition of smaller band-gap material such as HgS forms a quantum well around a CdS quantum dot. The quantum well traps rapidly the excited electron, which leads to a charge-separated state. Therefore, the quantum-dot-quantum-well system has a longer excited state lifetime than the bare CdS core. The addition of organic electron acceptors is another interesting technique to either speed up or retard the recombination of the photoexcited electron and hole. The different relaxation mechanisms of the chemically modified nanoparticles are compared. Our results have interesting implications for the use of such semiconductor nanoparticles in photochemical or photocatalytic applications.

2. **PHYS434** Femtosecond electronic absorption of vibrationally excited CH2I2 in solution: Watching the energy flow from bond to bond Andrew M. King, Dieter Bingemann, and F. Fleming Crim Jr., Department of Chemistry, University of Wisconsin, 1101 University Avenue, Madison, WI 53706, Fax: 608-262-9918, king@chem.wisc.edu Ultrafast transient absorption measurements on methylene iodide (CH2I2) in CCl4, CDCl3, and C6D6 after excitation of a C–H vibration allow direct observation of the time for intramolecular vibrational relaxation and energy transfer to the solvent. We excite the first overtone of the C–H stretching vibration in CH2I2 with a 100 fs laser pulse. Intramolecular energy redistribution populates vibrational states with larger Franck Condon factors for the electronic transition leading to an increased absorption of a second, time-delayed ultraviolet pulse, tuned over the wavelength range from 380 nm to 440 nm. We describe the transient absorption for all probe wavelengths with the temperature dependence of the absorption coefficient where the temperature varies exponentially as a function of time, reflecting the intramolecular and the intermolecular energy transfer. We obtain intramolecular vibrational relaxation times of 11 ps for the vCH = 2 state of CH2I2 in the solvents CCl4 and CDCl3, and a relaxation time of 8 ps in the solvent C6D6. The energy transfer to the solvent occurs with a time constant of 68 ps in CCl4, 51 ps in CDCl3, and 23 ps in C6D6.

3. **PHYS435** Friction on macroscopic and molecular timescales and chemical dynamics in liquids Steven A. Adelman, Department of Chemistry, Purdue University, 1393 Brown Building, West Lafayette, IN 47907-1393, Fax: 765-494-0239, and R. Ravi, Department of Chemical Engineering, IIT Kanpur Arrhenius' principle implies that liquid phase chemical reactions typically occur in a nonclassical "fast variable" near sudden limit timescale regime rather than in the "slow variable" near adiabatic limit regime of standard irreversible statistical mechanics. Despite this, the traditional theories of liquid phase reactions are of the slow variable type. For this reason a critical discussion of the standard

Langevin, Onsager, Mori slow variable model of irreversible dynamics is given which: (a) identifies the physical assumptions that underlie this model, and (b) points out the implications of these assumptions for the validity of the usual Kramers-type slow variable theories of liquid phase reaction rates. This discussion especially emphasizes that the standard model emerged from attempts to explain purely macroscopic phenomena and is ultimately founded on fully macroscopic measurements. Using the results of recent molecular dynamics simulations and ultrafast laser experiments, it shown that the macroscopic roots of the standard model significantly restricts its applicability to chemical reactions in liquids and other fast variable processes; since such processes are mediated by the subpicosecond timescale components of the solvent response. Finally a fast variable approach to short timescale and high frequency liquid phase processes is outlined; and its utility is illustrated using the results simulations and experiments.

4. **PHYS436** Isotope and temperature effect of proton transfer rate from extremely strong amine photoacids Tamar Barak1, Dina Pines2, and Ehud Pines2. (1) Department of Chemistry, Ben -Gurion University of the Negev, P.O.B. 653, Ber-Sheva 84105, Israel, Fax: 972-7-6472-943, epines@bgumail.bgu.ac.il, (2) Department of Chemistry, Ben-Gurion University We examine the proton transfer reaction of several primary, secondary and ternary amine photoacids. In particular protonated 1-aminopyrene trisulfonate (APTS) is an extremely strong photoacid with acidity comparable to that of the strongest known mineral acids.The proton transfer rate of amine photoacids is relatively slow up to 1011 s-1 in the case of APTS, exhibits a relatively large isotope effect kH/kD @ 5 and has a small activation energy Ea @ 2kcal/mol. This points out that specific amine-H20 hydrogen bonds interactions play an important role in the proton transfer process of protonated amine acids.

5. **PHYS437** Photophysical investigation of substituted cyclopropenylium cations: The smallest aromatic system Ling Zang, Xuejie Yang, Alex Gusev, Douglas C. Neckers, Wenqin Zhang, and Michael A. J. Rodgers, Department of Chemistry, Bowling Green State University, Bowling Green, OH 43403, Fax: 419-372-9809, lzang@bgnet.bgsu.edu As derivatives of the smallest Hückel (4n+2) aromatic system, triphenylcyclopropenylium (TPCP+) salts are stable in most polar organic solvents. The fluorescence lifetime is short (< 100 ps) as measured by a single photon counting method. This is consistent with the low fluorescence quantum yield (ca 2*10-3). Phosphorescence measurement at 77 K gives a triplet energy of 2.6 eV, 0.7 eV below the singlet energy. Appropriate anionic electron donors were chosen to compose ground state ion pairs with TPCP+, and photoinduced electron transfer involving the singlet state has been investigated by ultrafast transient absorption spectrometry. The spectra, compared to those from the putative intramolecular charge transfer state of diisopropylamino-substituted TPCP+ are expected to provide spectroscopic information about the TPCP neutral radical, which has not so far been characterized. Considering the high energy gap between the singlet and ground state, the very rapid deactivation of the singlet state is thought to occur through efficient intersystem crossing to the triplet state or another intermediate state. The possibility of diffusion controlled charge transfer quenching of the TPCP+ triplet state is being examined using a series of electron donors. These questions are currently under investigation using laser flash photolysis experiments at the ultrafast and suprananosecond timescales.

6. **PHYS438** Solar cell sensitizer dynamics and electron transfer Jeremy E. Monat1, Darius Kuciauskas2, Nathan S. Lewis2, and James K. McCusker1. (1) Department of Chemistry, University of California, Berkeley, CA 94720-1460, Fax: 510-642-8369, jemonat@socrates.berkeley.edu, (2) Division of Chemistry and Chemical Engineering, California Institute of Technology Ultrafast kinetic studies probing the electron transfer processes in TiO2-based solar cell sensitizers have been performed. Visible transient absorption (TA) experiments with a time resolution of ~100 fs on an iron(II) polypyridyl complex suggest that a long-lived charge-separated state is not necessary for electron injection into TiO2. Four sensitizers of the type MII(dcb)2X2 (M=Ru, Os; X=CN-, NCS-) have been

investigated on femtosecond and nanosecond time scales to study electron injection and recombination processes, respectively. A weak correlation has been observed between the driving force for electron injection and the picosecond dynamics of injection. On the other hand, the kinetics of electron recombination are strongly dependent on the identity of the sensitizer. Finally, the intramolecular dynamics of the dye RuII(dcb)2(NCS)2 have been studied in solution to determine the states from which subpicosecond and multipicosecond relaxation components occur.

7. **PHYS439** Unexpected signal in ultrafast gas-phase four-wave mixing Igor Pastirk, Bruna I. Grimberg, Vadim V. Lozovoy, and Marcos Dantus, Department of Chemistry, Michigan State University, East Lansing, MI 46624-1322, pastirki@msu.edu Experimental observation of four-wave mixing signals for pulse sequences that do not satisfy the rotating wave approximation are reported. The signals arise from the coupling between two molecular ensembles via a first-order free-induction decay emission. The solution of the Liouville equation for a four level system (two electronic states with two vibrations each) simulates the experimental results obtained for iodine in the gas phase.

8. **PHYS440** Coherent control through molecular phase manipulation in resonantly enhanced multi-photon dissociation-ionization of SO2 Hai-Lung Dai, Bin Xue, and Jun Han, Department of Chemistry, University of Pennsylvania, Philadelphia, PA 19104-6323, Fax: 215-898-2037, dai@sas.upenn.edu It is shown that molecular phase interference in coherent excitation can be used to manipulate molecular dissociation/ionization in time. Coherent excitation through a rovibronic transition in the C~X band of SO2, which displays fluorescence quantum beat, is used to gain access to either the low vibrational level (210) in the C state or the high vibrational level with 44877.52 cm-1 energy in the X state. The frequency of the second laser pulse is adjusted to induce resonance-enhanced excitation of SO2 through either the high vibrational level or the C state level wavefunction. The excitation results in dissociation into SO + O and ionization of the SO fragments. Depending on the resonance conditions, the ion signal is shown to be either in phase or 180* out of phase with the quantum beat.

9. **PHYS441** Excited state dynamics and rapid internal conversion in "stable dipoles" molecules Gavin D. Reid, Godfrey S. Beddard, Benjamin J. Whitaker, and Douglas Whittaker, School of Chemistry, University of Leeds, Woodhouse Lane, Leeds LS2 9JT, United Kingdom, Fax: 44-113-233-6565, g.d.reid@chem.leeds.ac.uk Femtosecond excited state dynamics in an ylide (C- -N+) 'stable dipole' molecule have been measured. Propagation of the wavepacket in the excited state towards the crossing region is recorded followed by ground state repopulation. Ab initio calculations suggest that a change in hybridisation of the nitrogen from SP2 to SP3 followed by buckling in the attached ring is responsible for the rapid internal conversion. These results are indicative of an essentially barrierless reaction and provide a new class of molecule with which to test theoretical models relating to dynamics near conical intersections.

10. **PHYS442** Femtochemistry of photochromic spirocompounds S. A. Antipin1, A. N. Petrukhin1, F. E. Gostev1, V. S. Marevtsev2, and O. M. Sarkisov1. (1) Department of Kinetics & Catalysis, Institute of Chemical Physics of Russian Academy of Sciences, 4, Kosygina street, 117977 Moscow, Russia, Fax: 095-938-2156, antipin@femto.chph.ras.ru, (2) Department of Polymers, Institute of Chemical Physics of Russian Academy of Sciences Process of photoisomerisation of nonsubstituted spironaphtooxazine, spironaphtopiran and spirophenantrooxazine was investigated by femtosecond "pump-probe" transient absorbtion spectral method. Dynamics of photoinduced states transient absorbtion spectra was registered. Supercontinuum was used as a probe pulse. Because of chirp of supercontinuum pulses all registered spectra were corrected by special method using pure toulene as standard substance. It was found that mechanism of photochemical reactions is the same for all compounds and includes the following stages: 1) excitation to S2 electronic state of closed form A; 2) relaxation to the excited vibrational levels of S1 state; 3) vibrational relaxation to the intermediate state X (C-O bond

cleavage); 4) relaxation from X to the ground electronic state S0 with formation of colored form B. All intermediates of this process were spectral observed. Rate constants of all stages were obtained. Difference in these values was explained by influence of compound structure.

11. **PHYS443** Femtochemistry of the paradigm NaI molecule in solution and clusters Gilles H. Peslherbe, Department of Chemistry and Biochemistry, Concordia University, 1455 DeMaisonneuve Blvd. West, Montreal, QC H3G 1M8, Canada, Fax: 514-848-2868, ghp@alcor.concordia.ca, Branka M. Ladanyi, Department of Chemistry, Colorado State University, and James T. Hynes, Département de Chimie, CNRS, Ecole Normale Supérieure Since the pioneering femtochemistry work of Zewail and coworkers, NaI has become the prototype for photodissociation dynamics involving covalent - ionic state curve crossing. Briefly, photoexcitation of ground state NaI to the first excited state results in bound oscillatory motion in the excited state potential, modulated by predissociation to the ground state. Our own interest in NaI centers on its photodissociation dynamics in solution and clusters. Our early theoretical work indicated that radiative deactivation to the ground ionic state would follow NaI ion pair photoexcitation in a weakly polar solvent, but with more interesting dynamics likely in small clusters. We have then simulated NaI photodissociation dynamics in polar solvent clusters with nonadiabatic MD techniques, focusing on mechanistic aspects and differences compared to isolated NaI. Clustered solvent molecules enhance the excited to ground state nonadiabatic transition probability, with the bound excited state showing muted oscillatory motion. The simulated photoionization probe signals agree with recent experiments.

12. **PHYS444** Femtosecond absorption spectroscopy of transition metal-based chromophores: Redefining the rules for inorganic photophysics James K. McCusker, Department of Chemistry, University of California, Berkeley, CA 94720-1460, Fax: 510-642-8369, mccusker@socrates.berkeley.edu Our research involves the application of femtosecond time-resolved absorption techniques to the study of the photophysics of transition metal complexes. In paricular, we are interested in exploring the dynamics of excited-state evolution in coordination complexes from their initially prepared Franck-Condon states to the lower energy excited states that have fueled much of the interest in the photo-induced properties of these compounds. This presentation will survey some of the results we have obtained concerning vibrational cooling dynamics, intersystem crossing processes, and the role of solvent in excited-state formation and thermalization in a range of chemical systems. The model that has begun to emerge from this work is at variance with that typically invoked for describing excited state dynamics in transition metal complexes, suggesting the need for an in-depth re-evaluation of the "rules" used for understanding the photophysics of inorganic chromophores.

13. **PHYS445** Femtosecond probing of photodissociation dynamics in acetyl cyanide Po-Yuan Cheng, I-Ren Lee, and Yu-Chieh Chung, Department of Chemistry, National Tsing Hua University, Hsinchu 30043, Taiwan, Fax: 886-3-571-1082, pycheng@mx.nthu.edu.tw Photodissociation of acetyl cyanide at ~ 6.4 eV energy region was studied by using femtosecond time-resolved laser-induced fluorescence (LIF) spectroscopy. Acetyl cyanide was excited by two-photon pumping at ~ 388 nm to the similar energy region reached by 193 nm one-photon excitation. A second delayed pulse, also at ~388 nm, then probed the formation of free CN radicals by monitoring the CN X --> B LIF signal. Our results showed that the temporal evolution of the CN(X) products can be well described by a bi-exponential rise function. The time constant for the fast component is about 2.5 psec and that for the slower one is about 25 psec. The fast component is about 9 times stronger in amplitude than the slower one. A dissociation mechanism has been proposed to explain the observed temporal behavior.

14. **PHYS447** Mechanism of the reaction, CH4 + O(1D2) ® CH3 + OH, studied by ultrafast and state-resolved photolysis/probe spectroscopy of the CH4·O3 van der Waals complex R. D. van Zee, C. C. Miller, and J. C. Stephenson, National Institute of Standards & Technology, 100 Bureau Drive, Gaithersburg, MD 20899, Fax: 301-975-3845, roger.vanzee@nist.gov The mechanism of the reaction

$CH_4 + O(^1D_2)$ ® $CH_3 + OH$ was investigated in state-resolved and time-resolved experiments. Ultraviolet pulses photolyzed ozone in the $CH_4 \cdot O_3$ van der Waals complex to produce $O(^1D_2)$. The ensuing reaction with CH_4 was monitored by laser-induced fluorescence through the OH A¬ X transition. In the state-resolved measurements, the distribution of OH(v=0,1; J; W; L) states, Pobs(v, J, W, L), was determined using a tunable, high-resolution laser. In the time-resolved measurements, an ultrafast laser system was used to monitor the appearance of these OH states at probe wavelengths centered between 307 nm - 316 nm. Because the ultrafast probe laser was spectrally broad, many rovibrational states were probed simultaneously. At each probe wavelength, multiple appearance rates were evident in the fluorescence signal, and the ratio of these components varied with probe wavelength. These data are most consistently fit using a three-mechanism model. The OH appearance signals, at all probe laser wavelengths, were best fit with time constants of tfast » 0.2 ps, tinter » 0.5 ps and tslow » 5.4 ps. The slowest of these three is the rate predicted by statistical theory for dissociation of a vibrationally excited methanol intermediate (CH_3OH^*) after complete intramolecular energy redistribution following insertion of $O(^1D_2)$ into CH_4. Under the assumption that the mechanism producing OH at the statistical rate would be characterized by a statistical prior, Pobs(v, J, W, L) was decomposed into three components, each with a linear surprisal. Dissociation of a CH_4O^* intermediate before complete energy randomization was identified as producing OH at the intermediate rate and was associated with a population distribution with more rovibrational energy than the slow mechanism. The third mechanism produces OH promptly with a cold rovibrational distribution, indicative of a collinear ion mechanism. From the decomposition of Pobs(v, J, W, L), it was possible to predict the fraction of signal associated with each mechanism at each probe wavelength in the ultrafast experiment, and the predictions proved consistent with measured appearance signals.

15. **PHYS448** Photoinduced dynamics in solid hydrogens Majed Chergui Sr., and Franco Vigliotti, Inst. de Physique de la Matière Condensée, University of Lausanne, Lausanne-Dorigny CH-1015, Switzerland, Fax: xx-41-21-692 36 35, Majed.chergui@ipmc.unil.ch The medium response of solid hydrogens to an impulsive perturbation has been studied using Rydberg excitation of an NO impurity. This leads to formation of an "electronic" bubble which corresponds to a cage radius increment of about 25%. The process has been followed in real-time using femtosecond pump-probe spectroscopy. Bubble dynamics is characterized by a one-way expansion of the cage with no recurrencies of the cage boundaries in both solid H_2 and D_2. The data is interpreted in terms of a dominant energy disspation by emission of sound waves and friction.

16. **PHYS449** Simulation of coherent nonadiabatic femtochemistry using classical trajectories Craig C. Martens, Department of Chemistry, University of California, Irvine, CA 92697-2025, Fax: 949-824-8571, cmartens@uci.edu The contributions by Ahmed Zewail to the study of chemistry on the femtosecond time scale, honored by the 1999 Nobel Prize in Chemistry, have opened a direct window on the fundamental steps of molecular processes and chemical transformation. In this talk, we describe our recent work on the development of theoretical methods for modeling and interpreting the view through this window. In particular, we present an approach to simulating the fully coherent dynamics of chemical systems on multiple coupled electronic surfaces using classical molecular dynamics. By employing a formalism based on a semiclassical limit of the exact multistate quantum Liouville equation, even manifestly quantum processes such as nonadiabatic wave packet interferometry—the interference of coherent multistate wavepackets undergoing nonadiabatic dynamics—can be treated classically. Quantum effects emerge in our approach as a breakdown of the independence of the members of the classical ensemble.

17. **PHYS450** Ultrafast energy redistribution in fullerenes Majed Chergui, Andreea Spiridon, Franco Vigliotti, Bernhard Lang, and Marcia Portella-Oberli, Inst. de Physique de la Matière Condensée, University of Lausanne, Lausanne-Dorigny CH-1015, Switzerland, Fax: xx-41-21-692 36 35,

Majed.chergui@ipmc.unil.ch The energy redistribution processes in C60 trapped in low temperature rare gas solids have been studied using one- and two-color femtosecond pump-probe spectroscopy. The data show an ultrafast internal conversion process, followed by a slower electronic relaxation among the lowest singlet states. That data are compared to results obtained in pure solid films of C60. They suggest that the intramolecular relaxation is also dominant in these later cases.

18. **PHYS451** Ultrafast nonradiative decay of electronically excited methyl viologen in water Jorge Peon, J. David Hoerner, Chungeng Xia, and Bern Kohler, Department of Chemistry, Ohio State University, 100 W. 18th Ave., Columbus, OH 43210, Fax: 614-292-1685, peon-peralta.1@osu.edu The photophysics of electronically excited bipyridinium dications are extremely sensitive to solvent. We have recently shown conclusively that these excited states fluoresce in nitrile solvents. In solvents of lower ionization potential, however, bipyridinium excited states show no radiative decay. Here we present the ultrafast dynamics seen in water. The results are discussed in terms of a dearomatization reaction recently proposed by Zewail and coworkers for the pyridinium cation. On the basis of our observed kinetic isotope effect, an alternative decay pathway via proton-coupled electron transfer is suggested.

19. **PHYS452** Nanoscale shock wave spectroscopy: A direct view of coherent ultrafast bath dynamics Craig C. Martens, and Daniela Kohen, Department of Chemistry, University of California, Irvine, Irvine, CA 92697-2025, Fax: 949-824-8571, cmartens@uci.edu We present molecular dynamics simulations of an idealized pump-probe experiment designed to permit the direct observation of the dynamical response of an impulsively-driven many-body bath in real time. A pump laser photodissociates an impurity molecule embedded in a solid host. This event initiates localized lattice disturbances—nanoscale shock waves—that travel through the lattice at supersonic velocities. The probe laser is tuned to a transient absorption in the host that is induced by the collisions of neighboring atoms that accompany the propagation of the local disturbance. The resulting pump-probe signal provides a direct view of the creation, evolution, and decay of the nanoscale shock waves. The general approach is applied to a model of molecular iodine embedded in a solid argon lattice, and the resulting synthetic signals are interpreted in terms of the microscopic dynamics of the system.

20. **PHYS453** Femtosecond dynamics of intramolecular electron transfer in photoexcited meso-tetraferrocenylporphyrins Victor A. Nadtochenko, Department of Kinetics and Catalysis, Institute of Chemical Physics Research of Russian Academy of Sciences, Institutskiy 14, Chernogolovka 142432, Russia, Fax: 007-096-515-35-88, nadto@icp.ac.ru, Oleg M. Sarkisov, Department of Kinetics, Institute of Chemical Physics of Russian Academy of Sciences, and Nikolai M. Loim, Institute of Organoelement Compounds of Russian Academy of Sciences The main goal of this work is to study femto-, picosecond relaxation dynamics of compounds: free base meso-tetraferrocenyl porphyrin(Fc4PH2), Zn meso-tetraferrocenyl porphyrin (ZnFc4P), diprotonated meso-tetraferrocenyl porphyrin (Fc4PH42+), EDA complex TCNE: Fc4PH2 in acetonitrile at room temperature These compounds can be considered as covalently linked donor -acceptor supramolecules and as nonplanar distorted porphyrins. Ferrocenyl substitute can be electron donor, Q1(*p-p) excited tetrapyrrole cycle can act as an electron acceptor. Experiments were performed by pump-supercontinuum-probe (PSCP) spectroscopy technique. The laser system is based on the CPM dye laser, multi-stage amplification. It produces pulses with a duration of 50 fs, centered at 610 nm with energy up to 100 mJ. Q1(*p-p)state was excited at l=610 nm . Differential spectra exhibit strong narrow bleaching band in the region of the appropriate B bands and wide absorption bands up to l=600 nm. Transients of the bleaching band exhibit fast 200-210 fs relaxation component in the case of Fc4PH2 and ZnFc4P, but this fast relaxation is not observed for the similar transients in the case of Fc4PH42+ and TCNE: Fc4PH2. In Fc4PH42+ and TCNE: Fc4PH2 the fastens component has a decay time about 10 ps. A detailed analysis of steady state absorption, fluorescence and 400 MHz proton NMR spectra allows to suggest: 1) Fc4PH2 and ZnFc4P exhibit fast conformational flexibility in the contrast to Fc4PH42+ and TCNE:

Fc4PH2, where the porphyrin plane inversion is slow in NMR time scale; 2) 200-210 fs relaxation component in Fc4PH2 , ZnFc4P and 10 ps relaxation component in Fc4PH42+ , TCNE: Fc4PH2 correspond to the transition from Q1(*p-p) state to charge transfer state Fc+-P-.

21. **PHYS454** Short time dynamics of alkali atoms on the surface of superfluid helium droplets: Ripplon excitation and desorption Frank Stienkemeier, Fakultät für Physik, Universität Bielefeld, Universitätsstr. 25, D-33615 Bielefeld, Germany, Fax: 011 (49) 521 106-6002, franks@physik.uni-bielefeld.de, and Claus Peter Schulz, Max-Born-Institut Berlin Superfluid helium nanodroplets are produced in a supersonic expansion from a cold nozzle and doped with alkali atoms and molecules. The metals are bound to the surface of the droplets. Femtosecond pump-probe spectroscopy is applied to study the dynamics of the systems: electronic excitation of potassium or cesium atoms induces strong repulsive interaction to the helium environment, the response of which can be followed in real time. We find the helium surface to relax within ca. 2ps into the new configuration. Attaching potassium dimers and trimers the desorption from the droplet can be followed and life times are extracted. Furthermore, observing the propagation of the induced wave packets, the vibrational motion of the dopant molecules can be analyzed. In this way information on the perturbation of the helium environment and its time dependence is obtained.

22. **PHYS455** Ultrafast spectroscopic studies of Coenzyme B12 derivatives and analogs Allwyn G. Cole, Neil Anderson, Joseph J. Shiang, and Roseanne J. Sension, Department of Chemistry, University of Michigan, 930 N. University, Ann Arbor, MI 48109, colea@umich.edu Ultrafast spectroscopy has been employed to monitor the photolytic cleavage of the cobalt-carbon bond in a variety of cobalamin analogs. Pump and probe pulses were generated in a noncollinear optical parametric amplifier. The pulses generated by the optical parametric amplifier were tunable between 470 nm and 690 nm and had pulse widths between 120 fs (unmodified) and 23 fs (recompressed). The cobalamin species studied here include the B12 coenzymes, 5'-deoxyadenosyl- and methylcobalamin, Vitamin B12 (cyanocobalamin), and the cobalamin analogs ethyl- and n-propyl-cobalamin. The excitation of 5'-deoxyadenosylcobalamin is wavelength independent and results in the homolytic cleavage of the cobalt-carbon bond analogous to the thermal cleavage that is observed in biological systems. On the other hand photolysis of methyl-cobalamin is wavelength dependent with a long-lived (1 ns) intermediate observed following excitation at 520 nm.

23. **PHYS456** Ultrafast studies of methyl sulfonyl unimolecular dissociation dynamics Jeffrey C. Owrutsky, and Andrew P. Baronavski, Chemistry Division, Naval Research Laboratory, Washington, DC 20375, Fax: 202-404-8119, jeff@chem1.nrl.navy.mil Ultrafast deep uv, mass-resolved photoionization spectroscopy has been used to study the unimolecular dissociation of methyl sulfonyl for the 193 nm photodissociation of methyl sulfonyl chloride (MSC) and methyl sulfonyl ethanol (MSE). In both cases, the parent excited state lifetime is instrument-limited (<200 fs). The decomposition lifetimes measured for methyl sulfonyl radical from MSC and MSE are 0.34 ps and 1.2 ps, respectively. The fractions of undissociated intermediate are <0.02 and 0.40, respectively. These results are remarkably well reproduced by RRKM calculations assuming an impulsive energy partitioning.

24. **PHYS457** Vibrational coherences: Coherent control and excitation dynamics Valeria D. Kleiman1, Dale McMorrow2, and Joseph S. Melinger2. (1) Electronics Sciences and Optical Technology Division, Naval Research Laboratory, 4555 Overlook Ave, Washington, DC 20375, valeriak@ccs.nrl.navy.mil, (2) Electronics Sciences and Optical technology Division, Naval Research Laboratory The manipulation of molecular dynamics using phase-modulated fields has experienced significant interest in recent years. One strategy used to control molecular reactions, is based on the creation of non-stationary wavepackets using ultrafast excitation pulses. The dynamics of the excited wavepackets can be manipulated by the use of characteristic phase-modulated fields in the excitation

process. Relatively few experiments have been performed on molecules in solution, where most chemical reactions occur. Recently, there has been a growing interest in utilizing ultrafast phase-modulation techniques to manipulate excitations in the condensed phase. Tailoring an excitation pulse so that a specific vibrational mode can be enhanced or suppressed is of vital interest when studying molecular systems in solution. We explore vibrational coherences in condensed phase molecules which are created by simple phase modulated femtosecond laser pulses. Experiments are performed using OHD-RIKES spectroscopy, which independently detects the real and imaginary components of the third order molecular response. The temporal evolution of the anisotropy was generated by the measurement of the probed beam as a function of the optical delay. Measurements were performed by detecting either the frequency-integrated probe pulse, or by dispersing the probe pulse and then detecting a narrow frequency band. For an excitation pulse with a specific duration and phase modulation, we find the experimental signals arising from the transient birefringence and dichroism to be qualitatively different.

25. **PHYS542** Femtosecond techniques to monitor and control chemical reactions on isolated molecules Thomas Baumert, Fachbereich Physik, Universität Kassel, Heinrich Plett Str. 40, D-34132 Kassel, Germany, Fax: ++49-561-804-4453, baumert@physik.uni-kassel.de As the femtosecond time scale is the time scale of nuclear motion within a molecule, femtosecond laser pulses can be used either to observe or to control molecular dynamics in real-time. The corresponding research area has been termed femtochemistry [1]. In this contribution we will first focus on femtosecond pump-probe photoelectron spectroscopy to study basic femtosecond coherent control schemes on the sodium dimer prototype [2][3]. The method is applied because of its sensitivity both to structural and electronic changes in a molecule upon excitation. Second we will discuss control of the branching ratios of different organometallic photodissociation reaction channels by feedback optimized phase-shaped femtosecond laser pulses [4]. [1] A. H. Zewail: Femtochemistry-Ultrafast Dynamics of the Chemical Bond I&II. New Jersey, Singapore: World Scientific, (1994). [2] A. Assion, M. Geisler, J. Helbing, V. Seyfried, T. Baumert: PRA 54, p.R4605 (1996). [3] T. Frohnmeyer, M. Hofmann, M. Strehle, T. Baumert: CPL 312, p.447, (1999). [4] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, G. Gerber: Science 282, p.919 (1998).

26. **PHYS543** Some interesting observed properties of metals confined in time and space of different shapes Mostafa A. El-Sayed, Laser Dynamics Lab, Georgia Institute of Technology, School of Chemistry and Biochemistry, Atlanta, GA 30332-0400, Fax: 404-894-0294, mostafa.el-sayed@chemistry.gatech.edu The type of electronic motion in matter determines its property and thus its uses in our everyday life. This motion itself is determined by the forces acting on the electrons which defines the space in which they are allowed to move. The difference between a metal, a semiconductor and an insulator lies in the fact that the electronic motion is highly delocalized, gently confined and highly confined, respectively. One thus expects that if we reduce the size of material to below its naturally allowed (God given) characteristic length scale new properties should be observed which are different from that of the macroscopic material as well as from those of their building blocks (atoms or molecules). This size is on the nanometer length scale. In this talk, I shall present some interesting new observations of the properties of metals when confined to this length scale.

27. **PHYS544** Ultrafast interferometry studies of light localization in photonic structures Yish-Hann Liau, and Norbert F Scherer, Department of Chemistry and James Franck Institute, University of Chicago, 5735 South Ellis Avenue, Chicago, IL 60637, Fax: 773-702-0805, nscherer@rainbow.uchicago.edu We report the first observation of pronounced recurrent signals in the pump-probe interferometry measurements of structured metallic interfaces. The coherent multiple scattering of surface plasmons from the micro-sphere over-layer causes light localization in a photonic structure.

Summary of Scientific Papers Presented

28. **PHYS545** Time-dependent electronic density matrix simulations of nonadiabatic photodynamics in molecules and aggregates Shaul Mukamel, Matteo Tommasini, Vladimir Chernyak, and Christoph Scheurer, Department of Chemistry, University of Rochester, P.O.Box 270216, Rochester, NY 14627-0216, Fax: 716-473-6889, mukamel@chem.rochester.edu A new time dependent method based on the Collective Electronic Oscillators (CEO) approach for computing excited state potential surfaces and non adiabatic couplings is used for simulating femtosecond nuclear dynamics in conjugated molecules and dendrimers. The algorithm avoids the prohibitively expensive direct computation of excited state many electron wavefunctions for many nuclear configurations along the trajectory. The CEO provide a fast and inexpensive algorithm for computing all these quantities. It is based on a reduced description of electronic structure which uses a minimal amount of relevant excited-state information and focuses on the time-dependent single-electron density matrix of the molecule. We have simulated pump-probe, time- and frequency-resolved fluorescence and three-pulse echo spectroscopies, of aggregates. The signals were recast using the Doorway-Window representation.

29. **PHYS546** Time-resolved dynamics in anion clusters using femtosecond photoelectron spectroscopy Daniel M. Neumark, Department of Chemistry, University of California, Berkeley, CA 94720, Fax: 510-642-6262, dan@radon.cchem.berkeley.edu Two experiments on time-resolved dynamics in size-selected cluster anions are performed using femtosecond photoelectron spectroscopy (FPES). In one experiment, vibrational relaxation dynamics are studied by coherent excitation of highly vibrationally excited I2- embedded in size-selected I2-(CO2)n clusters. FPES is then used to monitor the resulting dynamics as energy flows from the anion chromophore to the surrounding solvent molecules. In another experiment, electron solvation dynamics in anion clusters of the type I-(Sn) are probed via excitation of the charge-transfer-to-solvent band in which the excess electron on the iodide is ejected into the solvent network. The resulting solvent relaxation dynamics can then be followed using FPES.

30. **PHYS547** Femtosecond energy-transfer dynamics in iron-sulfur proteins Dongping Zhong, and Ahmed H. Zewail, Laboratory for Molecular Sciences, Arthur Amos Noyes Laboratory of Chemical Physics, California Institute of Technology, Pasadena, CA 91125, dongping@its.caltech.edu The fluorescence (and anisotropy) decay of the excited state of tryptophan has been studied both free in solution as well as in several proteins by femtosceond upconversion spectroscopy. Strong solvation dynamics of the free tryptophan were observed in ~1 ps. The resonance energy transfer, for example, between the intrinsic tryptophan residue and iron-sulfur in the rubredoxin occurs in as short as ~5 ps. The dependence of the energy-transfer rates on the initial molecular orientation, the hydrophobic interaction and the local nanoenvironment in proteins is discussed. The results are compared with the X-ray structures and those predicted from the Förster theory.

31. **PHYS548** Dynamic stark shifting of molecular energy levels by femtosecond laser pulses Rodrigo B. Lopez-Martens, Timothy W. Schmidt, and Gareth Roberts, Department of Chemistry, Cambridge University, Lensfield Road, CB2 1EW Cambridge, United Kingdom, Fax: 44-1223-336362, rl212@cus.cam.ac.uk, gr17@cus.cam.ac.uk The dynamic Stark effect in Rydberg NO induced by ultrafast laser pulses has been quantitatively determined over a range of incident laser intensities. Measurements were made by focussing frequency-doubled, sub-100 femtosecond pulses from an amplified Ti:sapphire laser into NO gas and monitoring the variation of A2S+ ® X2Pr fluorescence intensity as a function of peak laser intensity at photon energies scanned from non-resonantly below to non-resonantly above that for the NO A2S+ n'=2 ¬ X2Pr n"=0 two-photon transition in the weak field limit. Combined with precise temporal and spatial characterization of the incident laser pulses, this procedure allows a direct and simple measurement of the AC Stark effect, expressed as a fraction of the free electron ponderomotive energy: a dynamic shift of 0.36 eV is found at I=6 x 1013 W cm-2 and l=400 nm for example. These results can be quantitatively modelled by a numerically exact integration of the time-dependent Schrödinger equation for the Rydberg electron in an intense laser field and also by a simpler kinetic treatment of multiphoton absorption and ionization.

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32. **PHYS581** Femtochemistry: Some New Directions Ahmed H. Zewail, Laboratory for Molecular Sciences, Arthur Amos Noyes Laboratory of Chemical Physics, California Institute of Technology, Pasadena, CA 91125, Fax: 626-792-8456

33. **PHYS582** Probing wavepacket dynamics with femtosecond energy- and angle-resolved photoelectron spectroscopy Kazuo Takatsuka1, Yasuki Arasaki1, Kwanghsi Wang2, and Vincent McKoy2. (1) Department of Basic Science, Graduate School of Arts and Sciences, University of Tokyo, Komaba 3-8-1, Meguro-ku, Tokyo 153-8902, Japan, Fax: 81-3-5454-6588, kaztak@mns2.c.u-tokyo.ac.jp, (2) Laboratory for Molecular Sciences, Division of Chemistry and Chemical Engineering, California Institute of Technology, Mail Code 127-72, Caltech, Pasadena, CA 91125, Fax: 626-568-8824, mckoy@its.caltech.edu Recent studies have demonstrated how well suited femtosecond time-resolved photoelectron spectra are for mapping wavepacket dynamics in molecular systems. Theoretical studies of femtosecond photoelectron spectra which incorporate a robust description of the underlying photoionization dynamics should enhance the utility of such spectra as a probe of wavepackets and of the evolution of electronic structure. This should be particularly true in regions of avoided crossings, where the photoionization amplitudes and electronic structure may evolve rapidly with geometry. In this talk, we will present results of studies of energy- and angle-resolved femotosecond photoelectron spectra for wavepackets in the diatomic systems Na2 and NaI. Both cases involve motion through regions of avoided crossings. In Na2, however, wavepacket motion occurs on a single adiabatic potential with an inner and outer well and a barrier between them, while in NaI wavepackets move on the nonadiabatically coupled covalent (NaI) and ionic (Na+I-) potentials. These studies will be used to illustrate the insight into wavepacket dynamics that time-resolved photoelectron spectra provide. For example, in the case of NaI, angle-resolved spectra are seen to offer promise for probing real-time dynamics of intramolecular electron transfer occuring in the crossing region of the ionic and covalent states.

34. **PHYS583** Influence of hydration on the dynamics of proton transfer reactions A. Welford Castleman Jr., Daniel E. Folmer, Eric S. Wisniewski, Dennis A. Card, and Sean M. Hurley, Department of Chemistry, Penn State University, 152 Davey Lab, University Park, PA 16802, Fax: 814-865-5235, awc@psu.edu Proton transfer is one of the most ubiquitous processes in nature, and interest has prompted wide ranging studies in both the gas and condensed phase. A major theme of inquiry is elucidating differences between gas and condensed phase phenomenon, and studies in clusters are invaluable in this regard. The talk will focus on the dynamics of single and double proton transfer in 7-azaindole, including hydration effects on both the base-pair dimer and the hydrated monomer, and on proton transfer that takes place upon the solvation of acids, as revealed via femtosecond pump-probe spectroscopy. Findings from recent investigations further establish the role of clusters in effecting associated Coulomb explosion phenomenon observed in pump-probe experiments conducted at high laser fluence, and these related findings also will be presented.

35. **PHYS584** Extracting molecular interactions from observations of controlled quantum dynamics phenomena Herschel Rabitz, Department of Chemistry, Princeton University, Princeton, NJ 08544, Fax: 609-258-0967, hrabitz@princeton.edu The ability to control molecular motion using tailored laser pulses opens up the prospect of attaining dynamical information rich in content with regard to the underlying interatomic forces. Various algorithms will be discussed for extracting fundamental interactions from ultrafast high resolution imaging or other dynamics data.

36. **PHYS585** Initiating protein folding at the transition state M. Gruebele, Dept. of Chemistry, University of Illinois, 600 South Mathews Ave., Urbana, IL 61801, Fax: 217-244-0997, gruebele@scs.uiuc.edu

Summary of Scientific Papers Presented

37. **PHYS586** Spin-orbit relaxation and recombination dynamics in I2- and ICl- cluster ions: A new type of photofragment caging reaction W. Carl Lineberger, and Todd Sanford, JILA and Dept. of Chemistry and Biochemistry, University of Colorado, CB 440, Boulder, CO 80309-0440, Fax: 303-492-8994, wcl@jila.colorado.edu We report a new type of photofragment caging reaction, in which recombination is preceded by photofragment electronic relaxation. This rapid spin-orbit quenching occurs by solvent asymmetry mediated electron transfer. Photodissociation of ICl-(CO2)n clusters with a partially completed solvation shell affords insight as a consequence of the asymmetric charge distribution between the I and Cl atoms. Due to its smaller size and larger electron affinity, the Cl end of the diatomic anion will be preferentially solvated, leaving the I end almost bare for clusters with four or fewer CO2 solvents. Following photoexcitation to a repulsive state nominally correlating to an I- dissociation asymptote, we find Cl- -based products. The Cl--based products become the only products when the first solvent shell is completed. We briefly discuss this unexpected observation of no recombination in terms of intramolecular electron transfer and caging dynamics. Supported by NSF and AFOSR

38. **PHYS587** Ultrafast observation and control of molecular dynamics: Beyond the pump-probe method Marcos Dantus, Igor Pastirk, Vadim V. Lozovoy, and Bruna I. Grimberg, Department of Chemistry, Michigan State University, East Lansing, MI 46624-1322, Fax: 517-353-1793, dantus@msu.edu Our group has been exploring three-pulse four-wave mixing (FWM), a coherent nonlinear optical technique, with the goal of observing and controlling molecular dynamics. In contrast to the pump-probe method, the lasers are combined coherently; therefore, the pulse ordering can be used to control the sign of each laser interaction (action on the bra or ket). This, together with the time-dependent transition probability among the electronic states makes FWM a powerful platform for observation and control of molecular dynamics in ground and excited states. Results from our group will demonstrate control of rotational, vibrational and electronic degrees of freedom.

Principal Attendees

A. N. Petrukhin
A. Welford Castleman Jr.
Ahmed H. Zewai
Alex Gusev
Allwyn G. Cole
Andreea Spiridon
Andrew M. King
Andrew P. Baronavski
Benjamin J. Whitaker
Bern Kohler
Bernhard Lang
Bin Xue
Bruna I. Grimberg
C. C. Miller
Christoph Scheurer
Chungeng Xia
Clemens Burda
Craig C. Martens
Dale McMorrow
Daniel E. Folmer
Daniel M. Neumark
Daniela Kohen
Darius Kuciauskas
Dennis A. Card
Dieter Bingemann
Dina Pines
Dongping Zhong
Douglas C. Neckers
Douglas Whittaker
Ehud Pines
Eric S. Wisniewski
F. E. Gostev
F. Fleming Crim Jr
Franco Vigliotti
Frank Stienkemeier
Gareth Roberts
Gavin D. Reid
Gilles H. Peslherbe
Godfrey S. Beddard
Hai-Lung Dai
Herschel Rabitz
Igor Pastirk
I-Ren Lee
J. C. Stephenson
J. David Hoerner
James K. McCusker
Jeffrey C. Owrutsky
Jeremy E. Monat
Jorge Peon
Joseph J. Shiang
Joseph S. Melinger
Jun Han
Kazuo Takatsuka
Kwanghsi Wang
Ling Zang
M. Gruebele
Majed Chergui
Marcia Portella-Oberli
Marcos Dantus
Matteo Tommasini
Michael A. J. Rodgers
Mona Mohamed
Mostafa A. El-Sayed
Nathan S. Lewis
Neil Anderson
Norbert F Scherer
O. M. Sarkisov
Po-Yuan Cheng
R. D. van Zee
Reginald B. Little
Rodrigo B. Lopez-Martens
Roseanne J. Sension
S. A. Antipin
Sean M. Hurley
Shaul Mukamel
Stephan Link
Steven A. Adelman
Tamar Barak
Thomas Baumert
Timothy W. Schmidt
V. S. Marevtsev
Vadim V. Lozovoy
Valeria D. Kleiman
Victor A. Nadtochenko
Vincent McKoy
Vladimir Chernyak
W. Carl Lineberger
Wenqin Zhang
Xuejie Yang
Yasuki Arasaki
Yish-Hann Liau
Yu-Chieh